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LASER DETECTION OF VERY RARE LONG-LIVED RADIOACTIVE ISOTOPES

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INTRODUCTION

Laser spectroscopy techniques have made it possible to solve the cardinal problems of optical spectroscopy: (1) the spectral resolution of the Doppler-free nonlinear spectroscopy techniques has already reached a value of $R = \nu/\Delta\nu \simeq 10^{11}$ at a spectral resonance width of $\Delta\nu \simeq 10^3$Hz, the development of methods for further reduction of $\Delta\nu$ being under way;[1] (2) where femtosecond mode-locked tunable lasers are used, the time resolution amounts to a few tens of femtoseconds, i.e. only a few tens of light oscillation periods;[2] (3) the sensitivity of some techniques, atomic and molecular photoionization spectroscopy in particular,[3] reaches ultimate values - single atoms and molecules. There is one more, perhaps the last, problem of optical spectroscopy that is still to be solved; (4) high-selectivity detection of trace atoms and molecules in a real environment, particularly the detection of trace rare isotope atoms in the presence of an abundant isotope or the detection of trace molecules of a certain species in a molecular mixture. Subject to intensive development are now being various approaches that can combine a maximum possible sensitivity with an exceptionally high detection selectivity. The present lecture treats in short of some possible ways to solve the first of these problem - to attain a high selectivity of optical detection of very rare isotopes. This problem was introduced in Ref.[4] and discussed in Refs.[5,6].

1. RARE ISOTOPES AND EXISTING METHODS FOR THEIR DETECTION

There are a fairly large number of rare isotopes of cosmic origin, particularly those formed in the upper atmosphere as a result of nuclear reactions under of effect of cosmic rays. They include such isotopes as ^{10}Be resulting from interaction of galactic cosmic rays with the N and O nuclei, ^{14}C formed in the reaction between secondary neutrons and N, and ^{26}Al produced as a result of splitting of the Ar nucleus. These isotopes form in the upper atmosphere, precipitate, and accumulate on the earth's surface and ocean bottom. The rate of their precipitation in the ocean can be considered to remain constant during a long period of time exceeding their half-life $T_{1/2}$. There also exist some technogenic radioactive isotopes being a product of human activity, such as ^{85}Kr and ^{90}Sr. Table I lists some rare isotopes along with their half-lives and concentrations relative to the content of their main stable isotopes.

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Table 1. Some rare cosmogenic isotopes

Isotope	$T_{1/2}$, y	Relative concentration
^{10}Be	1.5×10^{6}	10^{-10}
^{14}C	5.7×10^{3}	$10^{-12} - 10^{-16}$
^{26}Al	7.4×10^{5}	10^{-14}
^{36}Cl	3.1×10^{5}	10^{-17}
^{41}Ca	8.0×10^{4}	10^{-21}
^{81}Kr	2.1×10^{5}	5×10^{-13}
^{85}Kr	10.8	10^{-11}
^{90}Sr	28.5	10^{-10}

The best known radioisotope among these is radiocarbon, ^{14}C, which is used for estimating the age of objects of organic origin.[7] Radiocarbon, which is formed in the upper atmosphere in a concentration of $^{14}C/^{12}C = 10^{-12}$, is involved in the Earth's biochemical life cycle. After an organism has ceased to be living and participating in the carbon cycle, its content of ^{14}C decreases exponentially in accordance with the 5730 - years' half-life of this radioisotope. To date organic archaeological objects or events of 50 000 years, for example, it is necessary to detect ^{14}C in relative concentrations a low as 10^{-15}. By utilizing other, longer-lived isotopes, radioisotope dating can be extended to cover millions of years in the past.

At present, there exist two universal methods for detecting cosmogenic radioactive isotopes in low concentrations. the first, most popular one consists in measuring the specific radioactivity of the sample under analysis and comparing it with that of a specimen of zero age. This standard quantity for ^{14}C, for example, is well known and amounts to 15.3 beta-decay events per minute per gram of the natural mixture of carbon isotopes. To realize this method requires fairly large samples (around 5 g) and a long observation time (approximately 1 day). Therefore, a major part of a very valuable sample has frequently to be sacrificed in order that its age can be determined. Serious measures should also be taken to ensure proper protection against background activity. The nuclear method of detecting rare isotopes is disadvantageous since it depends for its operation on radioactive transformations of the isotopes, which occur extremely seldom. Therefore, to have a reasonable observation time (a few days), the sample must, in principle, contain a large number of the rare isotope atoms of interest.

The second method for detecting rare isotopes consists in utilizing a linear[8] or cyclic[9] accelerator as a high-resolution mass spectrometer.[10] The principal difficulty in implementing this method is the ned to suppress background noise due to abundant isotopes and isobaric atoms such as ^{14}N in the case of ^{14}C. This problem can be solved for atoms having negative ions (C^-) and lacking negative ions of isobaric atoms (N^-). The method handles much smaller samples (down to 5 mg). The cost of the equipment required by the method is rather high.

It is clear that the shortcomings of the both generally accepted methods open up a wide field of application for laser techniques, for these are, in principle, capable of tackling the very difficult task of detecting a few rare isotope atoms against the background of 10^{10} to 10^{20} atoms of the most abundant isotope of the same atomic species.

2. POSSIBLE LASER TECHNIQUES. LIMITATIONS AND WAYS TO OVERCOME THEM

All the existing laser spectroscopy methods whose ultimate sensitivity lies at a level of single atoms[5,11] can, in principle, be employed to effect a highly selective detection of rare isotope atoms. Figure 1 presents simplified schemes of the three main laser techniques for detecting single atoms. These techniques take advantage of the effects due to resonant interaction of the atom with photons: (1) spontaneous reradiation of many photons absorbed from the laser beam; (2) photoionization of the atom as a result of absorption of few photons; (3) changes of the atomic coordinate and velocity consequent upon reradiation of a large number of photons.

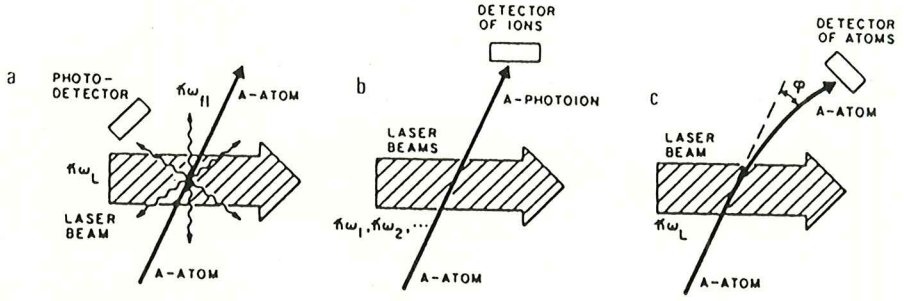

Figure 1. Various mechanisms of laser-atom interaction which can be used for atomic detection: (a) fluorescence; (b) photoionization; (c) deflection.

The main question relating to the applicability of these methods to detecting rare isotopes is their ultimate selectivity S, i.e. the ability to detect a small number (N_B) of the rare isotope atoms B in the presence of much greater number (N_A) of the main isotope atoms A:

$$S = N_B / N_A \qquad (1)$$

The selectivity of these techniques stems from the presence of a small isotope shift $\Delta = \omega_A - \omega_B$ of the spectral line of one or several consecutive resonant transitions of the atom from its ground state to an excited

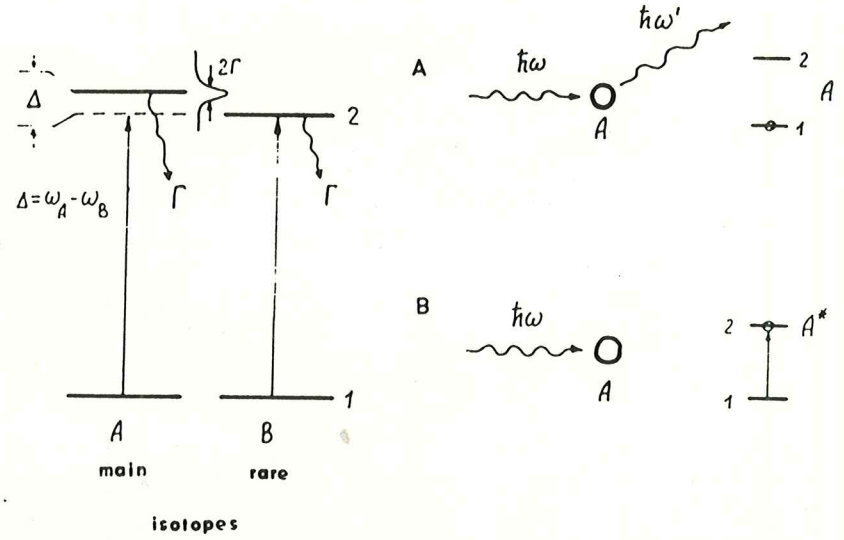

Figure 2. Limitation of detection selectivity if rare B atoms in presence of abundant A atoms with close spectral line: (a) method of fluorescent detection; (b) method of excitation detection.

state (Figure 2). The fact that the width of any spectral line is finite naturally limits the selectivity because of the overlapping of the wings of the close spectral lines of the atoms A and B, but the character of the limitation largely depends on the particular technique used.

With the fluorescence technique, the fluorescence excitation line usually has a Lorentzian shape, i.e. the probability that a photon will be scattered by the atoms A when the laser frequency coincides with the center of the spectral line of the rare atoms B is defined (in excitation conditions far from saturation) by the expression:

$$W^{A}_{scat} = \sigma_o I \mathcal{L}(\Delta\Gamma) \simeq (1/2)(\mu_{12}\mathcal{E}/\hbar\Delta)^2\Gamma \qquad (2)$$

where I is the laser radiation intensity (in photons/cm^{2}s), σ_o the cross section of the radiative transition $1 \longrightarrow 2$ at a maximum, Γ the natural half-width or the rate of spontaneous decay of the atom into its initial ground state, μ_{12} the dipole moment of the transition $1 \longrightarrow 2$, $\mathcal{E}$ the electric field strength of the light wave, and $\mathcal{L}(x) = 1/(1 + x^2)$ the Lorentzian function. By virtue of Eq.(2), the selectivity of the fluorescence detection of rare atoms is limited to the level of:

$$S_{fl} = W^{B}_{scat}/W^{A}_{scat} = (\Delta/\Gamma)^2 \qquad (\Delta \gg \Gamma) \ . \qquad (3)$$

For typical isotope shift and radiative linewidth values, $S_{fl} \simeq 10^4 - 10^6$, i.e. it is much lower than the required values indicated in Table 1.

The photoionization technique detects excited atoms by their subsequent transition into an ionized state. Its selectivity is therefore governed by the probability of the atomic excitation and not by that of photon reradiation. The probability of the atom A being excited on the wing of its spectral line is determined by the probability of absorption of two photons with the frequency $\omega = \omega_B$ and concurrent spontaneous reradiation of a photon with the shifted frequency $\omega_{fl} = 2\omega - \omega_A = \omega + \Delta$:[12]

$$W^{A}_{exc} \simeq (\mu_{12}\mathcal{E}/\hbar\Delta)^4\Gamma \propto \Delta^{-4} \ . \qquad (4)$$

This expression differs from Eq.(2), which is the one commonly used for estimation purposes, by a stronger dependence of the excitation rate on the frequency shift Δ, the difference being substantial, but the selectivity of the method depends materially on the type of subsequent ionization of the excited atoms A. In the case of nonresonant simultaneous ionization where the difference between the energy of the first absorbed photon and the atomic excitation energy, $\hbar\Delta$, can be compensated for by the second absorbed photon, the selectivity is reduced to the former level defined by Eq.(3). Where the atomic ionization is a resonance process or delays from excitation pulse, a higher selectivity can be attained.[12]

It follows from the above simple estimates that none of the laser techniques in its simplest version can provide for the very high detection selectivity required. However, each of the techniques can be modified so as to ensure a substantial increase in selectivity. The methods that are possible here may be divided into two groups:

(1) Methods based o n a repeated resonance interaction of the atom with the laser light, wherein the atom reradiates a large number of photons. In this case, there occurs what may be called a "selectivity accumulation" on account of an effect which is possible only where a single atom reradiates a large number of photons following its repeated resonance excitation. This effect is made use of in the method of "fluorescent bursts"[13,14] and that of laser deceleration and cooling of atoms.[15]

(2) Methods based on a multistep resonance excitation of the atom in a multiple-frequency laser field, wherein use is made of the isotope shift on several consecutive resonant transitions. As a result of such a multistep resonance excitation, the selectivities S_i attained at each excitation and ionization stage are multiplied.[16]

The method of "fluorescent bursts" can be used to design a photon burst atom counter at the inlets of mass-spectrometers or AMS machines in order to increase the selectivity and overcome isobar interferences.[17,18]

The creation of high-power single-frequency CW dye lasers has allowed resonance multistep excitation to be applied for isotope-selective ionization of atoms at the outlets of mass spectrometers.[19,20] To increase the detection selectivity the method of laser resonance depletion of the intermediate state was suggested[21] and realized.[22]

The idea of multiplication of isotopic selectivity at each excitation stage is difficult to realize for the most interesting rare isotopes (Table 1) because it is hard to find for them a series of consecutive upward transitions with noticeable isotope shifts, such shifts being characteristic of the ground state only.

3. METHOD OF MULTISTEP COLLINEAR PHOTOIONIZATION OF ACCELERATED ATOMS

A universal way to overcome this difficulty and make the method of multistep resonance photoionization really applicable to the detection of rare isotopes was suggested in [23]. The idea of the method is based on collinear stepwise photoionization of a beam of accelerated atoms. It is common knowledge that acceleration of atoms in the form of ions under a given potential difference U and subsequent neutralization of the ions into atoms lead to the bunching of the longitudinal ionic velocities, hence to the narrowing of the Doppler width of all the spectral lines of the given atomic species (if viewed in a collinear fashion) as compared with the initial Doppler width $\delta\nu_D(0)$ at an ion source temperature of T:[24]

$$\delta\nu_D(V)/\delta\nu_D(0) = (1/2)(kT/eU)^{1/2} \quad . \tag{5}$$

At $U = 10^4$ V the narrowing factor reaches 10^3. What is important is that the atoms in this case group in smaller volume of the phase space and their Doppler-free spectroscopy is effected without any loss of sensitivity.

Along with the narrowing of the Doppler width, there also occurs the Doppler shift of all the spectral lines of the accelerated atoms, which depends on the mass of the ion. As a result, there occurs an artificial "mass" shift on any spectral transition of the atom:

$$\delta\nu_{sh}/\nu_o = (1/c)(\sqrt{2eU})(1/\sqrt{M_1} - 1/\sqrt{M_2}) \quad . \tag{6}$$

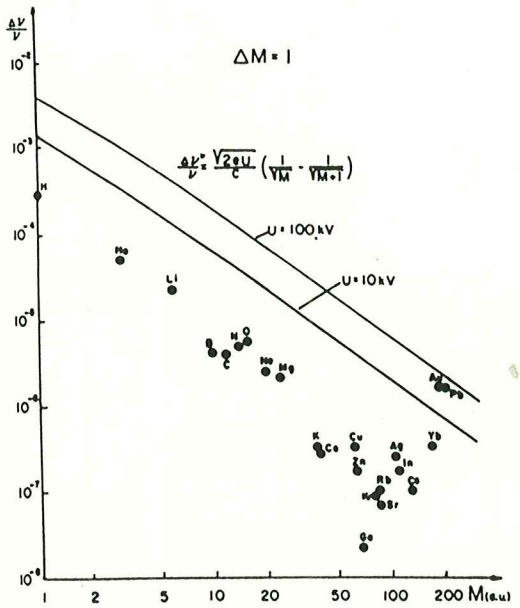

Figure 3. Values of relative isotope shift of resonance lines of various elements (dots). The kinematic isotope shift of accelerated atoms with energies 10 and 100 keV as a function of atomic mass (solid lines) for $\Delta M = 1$.

The circles in Fig.3 indicate the values of the natural isotope shift at resonant transitions of various elements and the artificial shift with the accelerating voltage. U = 10 kV and 100 kV. It can be seen that the kinematical isotopic shift may increase the isotope shift by 10 to 500 times. It is particularly important to apply the kinematic shift to elements in the middle of the periodic system.

What is most important is that such a kinematic isotope shift occurs on any spectral transition of the atom, this making it possible to realize in a natural way the idea of selectivity multiplication in multistep isotope-selective excitation.

Another important characteristic of this method is that it is possible to realize multistep isotope-selective excitation and ionization of elements with a high (~ 20 eV) ionization potential with the use of the available dye lasers. In this case the atoms should have high-lying metastable states.

General scheme of realization of this general idea is shown on Fig.4. A beam of fast atoms is formed by charge exchange of the ion beam with the atoms of the gas cell (alkali metal vapor, for instance). In the case of resonant charge exchange, when the ionization potential of the target atom is equal to the electron binding energy in the state, fast atoms are formed in the ground electronic state. In order to produce fast atoms with a high I.P. in the metastable state, one should choose such a gas cell that its ionization potential be close to the electron binding energy in this metastable state.

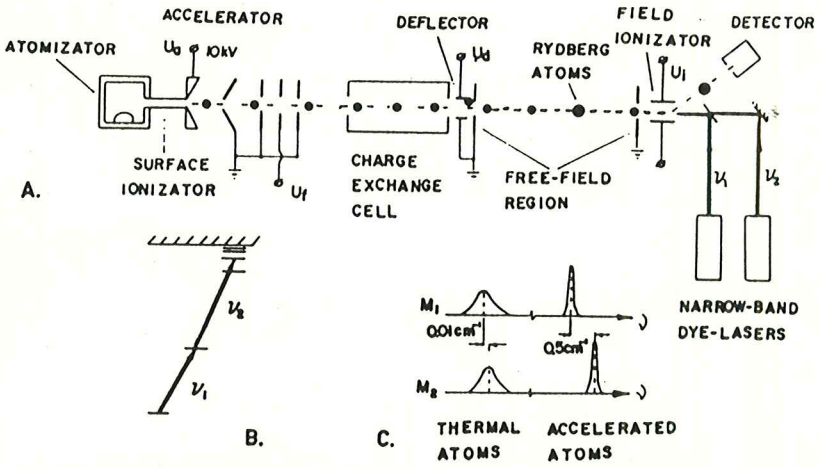

Figure 4. Laser detection of rare isotopes based on multistep collinear ionization.

4. EXPERIMENTAL DETECTION OF ^{40}K AND ^{3}He

The experiments on two-step collinear excitation of accelerated K isotopes to Rydberg states with their subsequent ionization in an electric field have proved the possibility of attaining the detection selectivity of the isotope ^{40}K equal to 10^5.[25] The isotope-selective excitation in this case was performed only at one step.

The K atom has two stable isotopes, ^{39}K and ^{41}K, with their relative concentrations of 93.2 and 6.8 %, respectively, and the radioactive isotope ^{40}K ($T_{1/2}$ = 1.3x10^9 years) with its relative concentration of 0.012 %. A beam of neutral K atoms was formed in the ground electronic state $4S_{1/2}$ by resonant charge exchange of accelerated K ions on K atoms. A beam of K ions was formed by ionizing the neutral atoms on the surface of tantalum foil heated to 1100 K. When the atoms are accelerated to an energy of 4 keV, the absorption line Doppler width of the transition $4P_{3/2}$ -$21D_{5/2}$ is reduced to $\delta\nu_D$ = 5.9 MHz.

The atoms were ionized by an electric field from the Rydberg state $21D_{3/2}$ upon two-step excitation at the transitions $4S_{3/1}$-$4P_{3/2}$, $4P_{3/2}$-$21D_{3/2}$ by laser radiation propagating to meet the atomic beam. The values of natural isotope shift at these transitions are 127 and 220 MHz, respectively. At the first transition the isotope shift is smaller than the hyperfine level splitting. After the atoms are accelerated to 4 keV, the absorption spectra of the isotopes are fully separated and the isotope shift increases to 2.5 GHz at the first transition and to 4.1 HHz at the second one.

In the experiments all the isotopes were excited at the transition $4S_{3/2}$-$4P_{3/2}$ by laser radiation with a spectral width of 0.8 cm^{-1}. the isotope-selective excitation of the atoms to Rydberg states was performed only at the second transition $4P_{3/2}$-$21D_{3/2}$ using a narrow-band laser with a spectral width of 0.015 cm^{-1}.

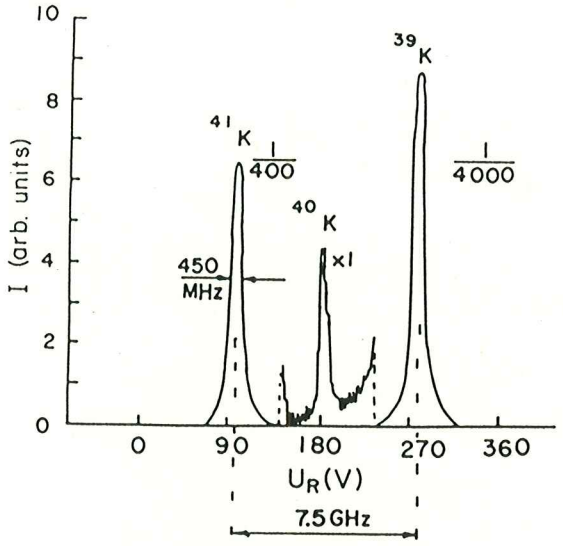

Figure 5. The ion signal of K isotopes as a function of detuning laser frequency and atomic resonance controlled by velocity variation of atoms.

The effect of multiplication of excitation selectivities at every step $S = S_1 \times S_2$ would be next natural experiment for a radical increase of the detection selectivity. Yet, it has been found, there is quite a number of elementary collisional processes to be taken into account in estimating the ultimate selectivity value.[26,27] The isotope ^{3}He, its natural concentration relative to the basic isotope ^{4}He being 10^{-6} was chosen to study these processes.

The ionization potential of the He atom E_1 = 24.6 eV, so the existing lasers cannot ensure multistep ionization from the ground state. The He atom has two long-lived metastable states": singlet 2^1S and triplet 2^3S. The charge exchange of accelerated ions on alkali metal vapor permits formation fast neutral atoms in metastable states. K atom is good candidate for such charge exchange. The electron binding energy of the states 2^1S and 2^3S of the He atom is close to the I.P. of K from the ground state $4S_{1/2}$. As a result of such charge exchange about 75 % of atoms are formed in the triplet metastable state 2^3S.[28]

The isotope-selective laser excitation of accelerated He atoms to the Rydberg n^3S and n^3D states with their subsequent ionization by an electric field was realized through the intermediate 3^3P level. To suppress this background noise, the method can be used in conjunction with various mass spectrometry techniques. in the present work, this was achieved through an additional time-of-flight separation of ^{4}He and ^{3}He, for which purpose use was made of an additional pulse intensity modulation of the continuous ion beam.[29] This made it possible to reduce the background noise due to $^4He^+$

ions by a factor of 10^4 and to detect ^{3}He with a relative abundance as low as 10^{-8}. This limit was set by the number of the ^{3}He atoms excited by the low-repetition-rate tunable dye lasers used in the experiment and not by the background noise.

Figures 6a and b illustrate the isotopic selectivity of the process of ionization of He atoms. Figure 6a shows the ion signal obtained while varying the frequency ν_2 of the second-step excitation laser, the first-step excitation laser frequency ν_1 = 25671.4 cm^{-1} being in resonance with ^{4}He (t_d = 5.2 μs). The background ions in the spectrum of ^{4}He are due to the two collisional processes.[29] The ion signal from ^{3}He with a relative content of 10^{-5} is shown in Fig.6b.

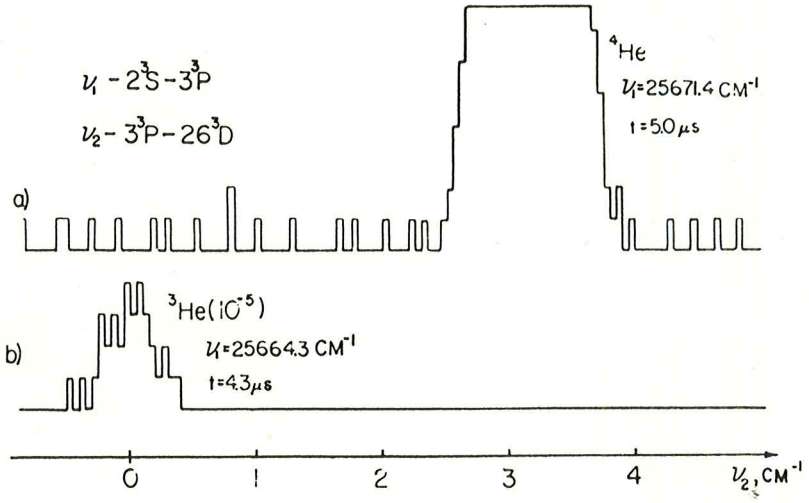

Figure 6. Ion signal as a function of the second-step excitation laser frequency ν_2. (a) The first-step laser frequency is in resonance with ^{4}He. (b) The first-step laser frequency is in resonance with ^{3}He. Averaged over 64 pulses.

The described technique for detecting the rare isotope ^{3}He, based on combining the collinear isotope-selective photoionization of fast atoms with the time-of-flight separation of isotopes, makes it was possible to measure the relative content of ^{3}He as low as 10^{-10}. The determination of the isotopic composition of helium is essential to the solution of many problems in geochemistry and geophysics, cosmochemistry, oceanology, and so on.[30] The isotopic ration [^{3}He]/[^{4}He] in various objects ranges between 10^{-4} and 10^{-10}. The method of collinear laser ionization of fast atoms can be used to solve these problems and to detect other rare isotopes as well.

5. POSSIBILITY OF DETECTION OF ^{81,85}Kr

Let us consider here the possibility of detecting ^{81,85}Kr isotopes by means o f laser collinear ionization in a fast atomic beam. As seen from Table 1, the detection of these isotopes must be realized with the selectivity $S = 10^{10}$-10^{12}. Such a selectivity can be provided in the case of two-step excitation by narrow-band laser radiation due to a kinematic isotope shift. The experiments on He isotope detection, however, show that the collisional processes of excitation and ionization of fast atomic beams on the molecules of residual gas represent a considerable limitation. The time-of-flight isotope separation due to atomic beam modulation cannot be used for such heavy elements as Kr . Yet it is rather convenient to apply collinear ionization in combination with pre-

separation in a mass separator. For this purpose, at the mass separator outlet the ions of the isotope to be detected separated from the basic isotope are directed into a charge exchange cell where they are transformed to atoms in the ground or the metastable states. If the attenuation of the line wing of the basic isotope on the mass of the isotope being detected due to mass separator $S_M = 10^4$, the spectral selectivity should be 10^6 to 10^8 to detect these isotopes.

The Kr atom has six stable isotopes: ^{86}Kr (17.3 %), ^{84}Kr (57 %), ^{83}Kr (11.5 %), ^{82}Kr (11.6 %), ^{80}Kr (2.25 %), and ^{78}Kr (0.35 %). The ionization potential of Kr I = 14 eV. Like in the case of He, the high-lying metastable states $1S_3$ or $1S_5$ can be used, too, as starting ones for laser multistep ionization. Pairs of alkali metals, such as K, Rb, and Cz, whose ionization potential is similar to the binding energy of electron in the state $1S_5$. The most effective transitions in the Kr atom where the radiation of dye lasers pumped by Cu lasers can be used. When the first-step radiation has a wavelength of 760.2 nm (the first-stage wavelengths are given for thermal atoms), the green line of Cu laser (λ_2 = 510.6 nm) can be used at the second step. The resonance tuning to the transition to a Rydberg state with n - 24,25 is realized by varying the atomic velocity. The nuclear spin of all the stable Kr isotopes, except ^{83}Kr, is equal to zero. The spectra o;f the rare isotopes ^{81}Kr (I = 7/2), ^{85}Kr (I = 9/2), and the stable isotope ^{83}Kr (I = 9/2) are characterized by a hyperfine structure. The magnitude of the natural isotope shift is about 50 MHz per one atomic mass[31] which is much smaller than that of hyperfine splitting.[32] The spectra of absorbed isotopes at the first stage (λ_1 = 788.5 nm) accelerated to an energy of 50 keV. The hyperfine splitting constants for this line are known only for the isotopes ^{83}Kr and ^{85}Kr. In this case the absorption spectra of the isotopes are fully separated. The magnitude of isotope shift of the most efficient component of ^{85}Kr is 3.7 GHz relative to ^{84}Kr and 1.4 GHz relative to ^{86}Kr. The radiative absorption linewidth is 6.12 MHz (τ_{rad} = 26 ns) and the laser excitation selectivity S_1 will be 2.1×10^5. The selectivity attained at the second excitation stage may be not less in magnitude.

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